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Impact of Soda Lime Aluminosilicate Glass Structure on Dissolution Kinetics in Simulated Cementitious Environment

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In order to sustain the rapidly increasing consumption of concrete, especially in developing countries, it is necessary to reduce the energy demands and CO_2 emissions associated with the production of Portland cement, are being investigated. Indeed, it is estimated that up to 10% of the global CO_2 emissions caused by human activities originates from the cement industry alone, where the CO_2 emissions are divided approximately evenly between the burning of fossil fuels and the calcination of limestone.[1)] Thus a partial replacement of Portland cement with a supplementary cementitious material (SCM) less rich in CaO has become an increasingly attractive approach to reduce this environmental issue.

A potential candidate for a SCM is calcium aluminosilicate glasses, which have similar chemical compositions of oxides as the main components in Portland cement, and they are relatively reactive in the strongly basic environment of the pore solution in hydrated cement.[2)] Ideally, waste glasses (cullet) from other industries could be utilized as SCMs, but the required amount to have a substantial impact is simply too large compared to the availability of cullet in most of the world. Thus, in this work we investigate the suitability of synthesized calcium aluminosilicate glasses as SCM by considering the relation between the glass structure and the dissolution and reactivity of the glasses in a simulated cementitious environment. The objects of this study are a multiple series of calcium aluminosilicate glasses, each modified to various degrees by alkali oxides.

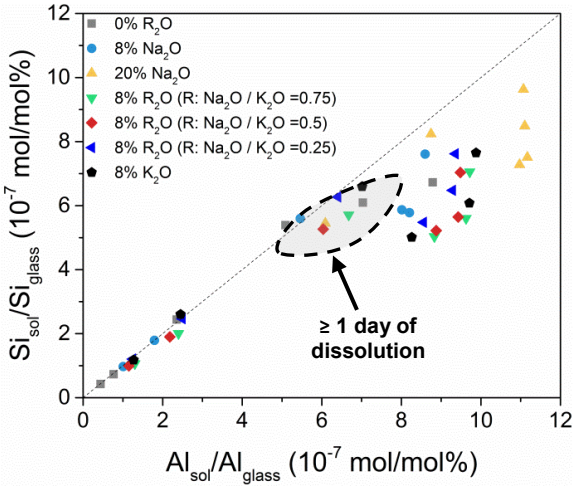

Figure 1. Si vs. Al in solution normalized to Si and Al in the dissolved sample, respectively. Note glasses with ≠ 8% alkali oxides show different kinetics than glasses = 8% alkali oxides. All glasses have similar Si/Al/Ca ratios.

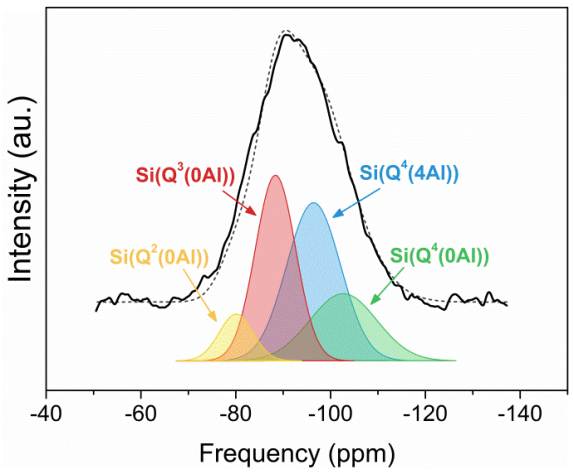

Figure 2 Example of Si(Q^n(kAl)) unit distribution in glass, simulated (dashed, black line) from 29Si MAS NMR spectrum of glass sample (solid, black line).

As seen in Fig. 1, a certain series of glasses show congruent dissolution until a certain point in time, depending on alkali content. This initial, seemingly diffusion controlled process thus determines the availability of ions in solution at early stages of hydration of glass particles. However, the kinetics of the individual glass dissolutions may be understood in terms of the glass structure, and the latter is explored by ^{29}Si MAS NMR spectroscopy and simulating the Si(Q^n(kAl)) unit distribution as illustrated in Fig. 2. By taking into account the degree of aluminum avoidance, the coordination of Si, Al, Ca, Na, and K is discussed in relation to the reactivity of the glasses.

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2) M. Moesgaard, D. Herfort, J. Skibsted, Y.Z. Yue, Calcium aluminosilicate glasses as supplementary cementitious materials, *Glass Technology – European Journal of Glass Science and Technology Part A* **51** (2010) 183-190.